Science of the Total Environment 619–620 (2018) 854–865

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Multi-contamination (heavy metals, polychlorinated biphenyls and polycyclic aromatic hydrocarbons) of littoral sediments and the associated ecological risk assessment in a large lake in France (Lake Bourget)

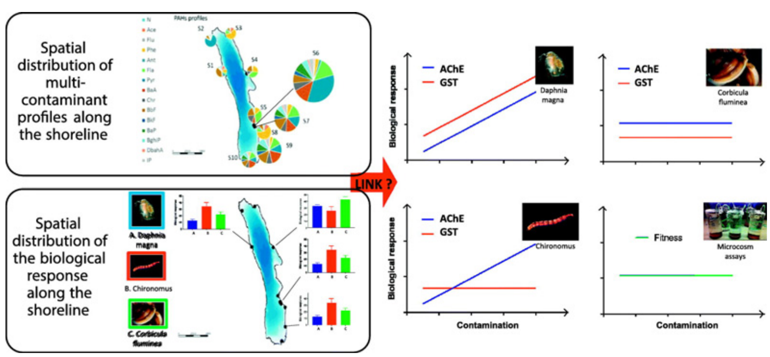
Nathalie Lécrivain [a,*], Vincent Aurenche [a], Nathalie Cottin [b], Victor Frossard [c], Bernard Clément [a]

[a] Laboratoire d'Ecologie des Hydrosystèmes Naturels et Anthropisés (LEHNA), UMR CNRS 5023, ENTPE, 69120 Vaulx-en-Velin, France
[b] Université de Savoie Mont-Blanc, LCME, 73376 Le Bourget du Lac, France
[c] Université de Savoie Mont-Blanc, UMR 42 CARRTEL, 73376 Le Bourget du Lac, France

HIGHLIGHTS

- Contamination showed spatial heterogeneity in the littoral of a large French lake.
- The laboratory and field biological responses spatially differed along the littoral.
- Link between chemical and biological status depended on study endpoint and organism.
- Littoral compartment should be considered when assessing lake ecological risk.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 15 September 2017
Received in revised form 13 November 2017
Accepted 14 November 2017
Available online xxxx

Keywords:
Pollutant mixture
Microcosm assays
Biomarker
Trace metallic elements
PAHs
PCBs

ABSTRACT

The lake littoral sediment is exposed to a large array of contaminants that can exhibit significant spatial variability and challenge our ability to assess contamination at lake scale. In this study, littoral sediment contamination was characterized among ten different sites in a large peri-alpine lake (Lake Bourget) regarding three groups of contaminants: 6 heavy metals, 15 polycyclic aromatic hydrocarbons and 7 polychlorinated biphenyls. The contamination profiles significantly varied among sites and differed from those previously reported for the deepest zone of the lake. An integrative approach including chemical and biological analyses was conducted to relate site contamination to ecological risk. The chemical approach consisted in mean PEC quotient calculation (average of the ratios of the contaminants concentration to their corresponding Probable Effect Concentration values) and revealed a low and heterogeneous toxicity of the contaminant mixture along the littoral. Biological analysis including both laboratory (microcosm assays) and in situ (Acetylcholine Esterase (AChE) and Glutathione S-Transferase (GST) activity measurements) experiments highlighted significant differences among sites both in the field and in laboratory assays suggesting a spatial variation of the biota response to contamination. Linear regressions were performed between mean PEC quotients and biological results to assess whether littoral ecological risk was explained by the contamination profiles. The results highly depended on the study benthic or pelagic compartment. Regarding autochthonous *Corbicula fluminea*, no significant relationship between mean PEC quotients and biomarker activity was found while a significant increase in AChE was observed on

* Corresponding author.
E-mail address: nathalie.lecrivain@entpe.fr (N. Lécrivain).

http://dx.doi.org/10.1016/j.scitotenv.2017.11.151

autochthonous chironomids, suggesting different stress among benthic organisms. Both AChE and GST in caged pelagic *Daphnia magna* showed a significant positive relationship with mean PEC quotients. This study underlines the importance of accounting for spatial variations in lake littoral sediment contamination and the need for performing an integrative approach coupling chemical, field and laboratory analyses to assess the ecological risk.

1. Introduction

Chemical contamination is a major threat on ecosystems worldwide (Bernhardt et al., 2017). As receiving environments, freshwaters and especially lakes collect and accumulate a large array of contaminants that can significantly alter their health (Malaj et al., 2011). Chemical contamination has traditionally been assessed by considering deep sediment (Roussiez et al., 2006). Despite the fact that these deep sediments can efficiently show contamination and are suitable for determining the historical pollution of lakes (Fernández et al., 2000; Naffrechoux et al., 2015), they may not be representative of the spatial heterogeneity of the contamination at the lake scale. Spatial variations of contaminant contents have been measured at the global scale (Marvin et al., 2004; Suresh et al., 2012). Such spatial variations are expected to occur in the littoral zone in particular because of its proximity to local sources of pollution inputs (Roussiez et al., 2006). Consequently, contamination in lake littoral sediments consists of a complex mixture of numerous chemicals reflecting the multiplicity of point sources and inducing differential ecological risks along the shoreline.

From this perspective, a widely used approach combining chemical screening, ecotoxicological tests and ecological surveys named the Triad approach, which was first proposed by Long and Chapman (1985), has been shown to provide robust assessments of ecological risk (Chapman et al., 2002; De Castro-Català et al., 2016; Marziali et al., 2017). Regarding the chemical approach, relevant tools can be used to compare the measured total content of individual chemicals in the sediment to ecotoxicological thresholds from the literature (e.g., the toxic unit approach, the comparison with consensus-based thresholds) (Höss et al., 2011; Long, 2006; MacDonald et al., 2011). However, since contaminants are present in various geochemical forms, bioassays are more appropriate to assess the bioavailable fraction of chemicals. Bioassays are usually conducted in a laboratory (e.g., microcosm assays) to study endpoints from the community to the sub-cellular level (Triffault-Bouchet et al., 2005). Additionally, in situ exposures can be performed to accurately observe the biological response in the conditions under which the fauna are exposed. Stress measurements can also be studied at a sub-cellular level using enzymatic biomarkers to provide information about the effects of contaminants (Capela et al., 2016). Among these, Acetylcholinesterase (AChE) is largely used as a biomarker of exposure to neurotoxic chemicals (Matozzo et al., 2005). It has been demonstrated that AChE activity in aquatic benthic invertebrates is mainly inhibited by carbamates (Alves et al., 2002) and organophosphorus compounds (Doran et al., 2001). However, as many other compounds affect the neurotoxicity of organisms, AChE activity can be impacted by other chemicals, such as heavy metals and persistent organic pollutants (Devi and Fingerman, 1995; Diamantino et al., 2000; Frasco et al., 2005; Guilhermino et al., 1998; Payne et al., 1996; Tilton et al., 2011). Another commonly studied biomarker is Glutathione S-transferase (GST), the activity of which is used for the detoxification of alkylating agents. An increase in this activity can thus be seen as a response to an oxidative stress following exposure to toxic compounds (Habig et al., 1974).

Lake Bourget is the largest natural lake in France. Its preservation is of great concern for economic reasons (tourism, drinking water resource), and the ecological status of the lake has thus been largely studied (Berdjeb et al., 2011; Jacquet et al., 2005). Past studies have observed the presence of several contaminant families, such as heavy metals and organic pollutants (PAHs, PCBs), in offshore sediments (Jung, 2009; Jung et al., 2008; Naffrechoux et al., 2015). In this study, we aimed to assess the spatial variability at the littoral scale of this contamination and the associated ecological risk by using an integrative chemical and biological approach, including laboratory microcosms and biomarker (AChE and GST) activity measurements. By comparing these results to historical data obtained from the deep zone, this work intends to provide the first contribution to comparing littoral versus offshore sediment analyses.

2. Materials and methods

2.1. Study sites

Lake Bourget is located in Savoy, on the northwest edge of the French Alps (45°43′N, 5°52′E, Fig. 1), and is the largest natural lake in France with a surface area of 42 km^2. The average and maximum depths are approximately 81 m and 145 m, respectively. Lake Bourget currently has an oligo to meso-trophic state. The 560 km^2 catchment area of Lake Bourget is covered by forests (45%), agricultural areas (22%), grazing fields (12%), urban areas (13%) and water bodies (8%) (Lake Bourget excluded) (Cisalb, 2009). Over the 61 municipalities of this catchment area, Chambéry and Aix-les-Bains are the two largest cities, with a combined population of 90,000 inhabitants. The Leysse, Tillet and Sierroz Rivers are the main lake tributaries. They both flow through urban, industrialized and rural areas. Specifically, the Leysse River runs through Chambéry city, and both the Tillet and Sierroz Rivers flow through the Aix-les-Bains conurbation. The north and northwest areas of the lake are less urbanized than the south and south-eastern areas, where the two largest cities are located. Forestry is predominant in the north, and steep-slope areas in the west limit urban expansion. Lake Bourget is bordered by a railway and roads from south to north along the eastern shore, and the Chambéry airport is located on the southern side of the lake, which may contribute to the littoral contamination.

The sediment pollution in Lake Bourget was already investigated in terms of offshore sediments (Arnaud et al., 2006, 2004; Chalhoub et al., 2013; Herve, 1998; Jung et al., 2008, 2007; Naffrechoux et al., 2015) and those close to a tributary outlet (Chalhoub et al., 2013) (Table 1 and Fig. 1C).

Ten sites were selected along the littoral zone of the lake for sediment characterization (S1 to S10, Fig. 1C and Table S1). They were chosen according to the anthropogenic activities located on the shoreline or at the exit of the main tributaries. S1 was selected in a pristine area, where no point source was specifically identified, and S2, S3, S8 and S9 at nearby beaches. S5, S7, S9 and S10 are located close to a river outlet. S4 is adjacent to a railway and S6 to a car park. Six of these ten sites (S2, S3, S6, S8, S9 and S10) were selected for biomarker analysis on autochthonous benthic macro-invertebrates and for exposure to caged laboratory *Daphnia magna*. These six sites were selected according to the large array of contamination intensity they exhibit and because they provided enough autochthonous macro-invertebrates for further analysis.

2.2. Sampling

At each site, surface (0–10 cm) sediment was collected at a water depth of 1.5 to 2.5 m using an Eckman grab. The sediment was then